Developing interface localized liquid dielectrophoresis for optical applications

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ABSTRACT

Electrowetting charges the solid-liquid interface to change the contact area of a droplet of a conducting liquid. It is a powerful technique used to create variable focus liquid lenses, electronic paper and other devices, but it depends upon ions within the liquid. Liquid dielectrophoresis (L-DEP) is a bulk force acting on the dipoles throughout a dielectric liquid and is not normally considered to be a localized effect acting at the interface between the liquid and a solid or other fluid. In this work, we show theoretically how non-uniform electric fields generated by interdigitated electrodes can effectively convert L-DEP into an interface localized form. We show that for droplets of sufficient thickness, the change in the cosine of the contact angle is proportional to the square of the applied voltage and so obeys a similar equation to that for electrowetting – this we call dielectrowetting. However, a major difference to electrowetting is that the strength of the effect is controlled by the electrode spacing and the liquid permittivity rather than the properties of an insulator in a sandwich structure. Experimentally, we show that that this dielectrowetting equation accurately describes the contact angle of a droplet of oil viewed across parallel interdigitated electrodes. Importantly, the induced spreading can be complete, such that contact angle saturation does not occur. We then show that for thin films, L-DEP can shape the liquid-air interface creating a spatially periodic wrinkle and that such a wrinkle can be used to create a voltage programmable phase diffraction grating.

Keywords: electrowetting, dielectrophoresis, liquid lens, contact angle, wetting, dielectrowetting, L-DEP, optofluidics.

1. INTRODUCTION

How liquids wet solid surfaces is of fundamental importance to a wide-range of applications from creating thin films on semiconductor wafers, through adhesion and coating of surfaces, to effective droplet deposition and mixing on DNA microarrays. The wetting of solid surfaces can be modified by changing material or surface properties, such as the surface chemistry or micro- or nano-scale topography[1], or by introducing additional energies, such as electrostatic[2,3]. In 1993, Berge recognized that electrostatic fields could be a method to create reversible hydrophilicity on a hydrophobic surface[4,5], and that this was a powerful method to control and manipulate droplets of liquid. Since that time the field of electrowetting-on-dielectric ("electrowetting") has grown from less than 20 journal articles per year to over 170 last year with 4000 citations annually (source: http://wok.mimas.ac.uk/). This growth has been due to its significance across diverse scientific disciplines with applications in optical devices[6,7], displays[8] and microfluidics[9,10]. During this time, major electrowetting-based international companies have been founded with examples being Varioptic (variable-focus liquid lenses), Liquavista (electrowetting-based displays) and Advanced Liquid Logic (digital microfluidics). All electrowetting work uses the concept that the contact area of a conducting liquid, typically a salt solution, can provide one contact in a capacitive structure where an electrode on a substrate is separated from the conducting liquid by a (hydrophobic) dielectric film. This allows a reversible charging of the solid-liquid interface which thereby reduces the contact angle allowing the radius of curvature of the droplet to be adjusted or droplet motion to be actuated. Electrowetting can be used in air or in a second immersion fluid and can involve AC or DC voltages.

In electrowetting the energy balance at the three-phase contact line is modified by a capacitive energy resulting in a contact angle which decreases with applied voltage. Modifying contact angles differently around the droplet periphery can be used to actuate contact line motion. Whilst electrowetting is a versatile method of controlling contact angle, traditionally it uses a conducting liquid in contact with an electrode and when operated with a droplet in air often has a high degree of contact angle hysteresis. It also suffers from a saturation effect that prevents the lowest contact angles and

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films being achieved. Electrowetting is one example of the wider effects of electromechanics as recognized by Jones et al[11-14]. When a dielectric liquid experiences a non-uniform, rather than uniform, electric field a bulk liquid dielectrophoretic force occurs that can actuate the motion of the liquid. Intuitively, two ends of a dipole experiencing slightly different magnitudes of electric fields from a non-uniform field will produce forces that do not cancel; therefore a net force occurs in a non-conducting liquid (or a conducting liquid using a high frequency electric field). Bulk liquid dielectrophoresis (L-DEP) has been used to drive liquid motion and to create dielectric liquid lenses[11,15-17], but this liquid dielectrophoresis effect is not usually thought of in terms of an interface localized force driving wetting phenomena.

In this paper, we present an approach to the wetting of solids by non-conducting dielectric liquids in a non-uniform decaying electric field that uses an interface localized form of liquid dielectrophoresis where the energy changes can be viewed as occurring at the interfaces between the solid, liquid and vapor phases. We discuss three regimes. The first focuses on droplet spreading and control of contact angles. In this case, the L-DEP energy is linked to changes of the solid-vapor and solid-liquid interfacial areas; an effect we call "dielectrowetting". The second focuses on films and corresponds to a far-field regime in which the L-DEP energy is linked to changes in shape of the liquid-vapor interface. The third regime also focuses on films, but corresponds to a near-field regime where the shape of the liquid-vapor interface reflects detailed variations in the electric field close to its source. We review our recent results showing that the droplet regime has a contact angle that obeys an equation similar to the electrowetting equation, but with the strength of the effect controlled by the relative permittivity of the liquid and the penetration depth of the electric field into the liquid[18]; this provides an understanding of possible parameters in the design of voltage controllable lenses driven by liquid dielectrophoresis. We also describe an application of these ideas to the film regime to create a voltage programmable amplitude phase grating[19].

2. CONCEPTUAL BASIS OF DIELECTROWETTING

2.1 Liquid dielectrophoresis and solid-liquid/liquid-fluid interfaces

A simple view of a dipole in a uniform electric field is that the forces at each end are equal in magnitude, but opposite in direction due to the opposite signs on the charges at each end of the dipole; there is therefore no net force. However, when the field is non-uniform a net force arises because of the difference of the magnitude of the electric field at the two ends of the dipole[20]. Now consider a uniform layer of a dielectric liquid of depth h on a solid surface with an electric potential that decays with depth of penetration into the liquid, i.e. $V(z)=V_o\exp(-2z/\delta)$, where δ is a penetration depth. The electrostatic energy per unit contact area, w_E, stored in the liquid is found by integrating the dielectric energy density, $\frac{1}{2}\varepsilon_o\varepsilon_l\underline{E}.\underline{E}$, where ε_l is the dielectric constant of the liquid and $\underline{E}=-\underline{\nabla}V$ is the electric field, over the volume of the liquid[18,19,21],

$$w_E = -\frac{\varepsilon_o\varepsilon_l V_o^2}{2\delta}\left[\exp\left(\frac{-4h}{\delta}\right)-1\right] \tag{1}$$

The exponential decay of the electric potential results in three regimes. In the first of these the liquid layer is sufficiently thick ($h>>\delta$) that the upper surface of the layer of liquid has no influence and the dielectrophoretic energy per unit contact area, and eq. (2) becomes $w_E=\varepsilon_o\varepsilon_l V_o^2/2\delta$. Any change in the solid-liquid interfacial area therefore involves a proportional change in the dielectrophoretic energy; effectively the effects of L-DEP are localized to the solid-liquid (and solid-vapor) interface. In the second regime, the liquid layer is sufficiently thin that the electric potential is still of significant value at the upper surface of the layer of liquid. In this case, any change in the shape of the liquid-vapor interface results in changes of the dielectrophoretic energy; this argument can be extended to a liquid-liquid interface in a two-liquid system. Typically, liquid gathers into regions of highest electric field gradient. This thin film regime can be further thought of as two regimes depending on whether the upper surface of the layer of liquid is influenced by an average far-field or by a detailed near-field of the non-uniform electric field close to its source. Thus, whilst L-DEP is, in principle, a bulk effect, its affects can also be considered in terms of how its energy contribution balances against interfacial energies as changes in interfacial areas occur.

2.2 Droplet regime

For a droplet, the wetting of a solid surface is given by a local minimum in the surface free energy arising from the solid-vapor, solid-liquid and liquid-vapor interfaces. These are characterized by three interfacial tensions, γ_{SV}, γ_{SL} and γ_{LV} (the

surface energies per unit area). Considering Fig. 1, the effect of increasing the contact area by a small amount, ΔA, is to replace the solid-vapor interface by a solid-liquid interface and so change the surface free energy by $(\gamma_{SL}-\gamma_{SV})\Delta A$.

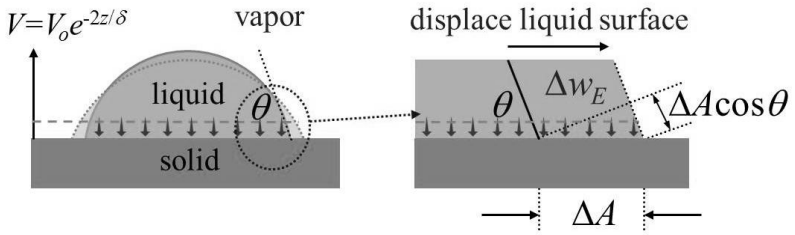

Figure 1. Energy changes for a thick droplet in an electric field decaying exponentially with distance from the substrate.

In addition, the movement of the liquid creates an additional liquid-vapor surface area of $\Delta A\cos\theta$ resulting in a surface free energy increase of $\gamma_{LV}\ \Delta A\cos\theta$.[22] In the presence of the non-uniform electric field, and assuming the droplet is sufficiently thick ($h>>\delta$), the L-DEP energy changes from its value in the vapor to that in the liquid $-\varepsilon_o(\varepsilon_l-1)\ V_o^2\ \Delta A/2\delta$. For the droplet to be in equilibrium the total change in energy must vanish and so the contact angle adopts a value, $\theta_e(V_o)$, given by[18],

$$\cos\theta_e(V_o)=\cos\theta_Y+\frac{\varepsilon_o(\varepsilon_l-1)V_o^2}{2\gamma_{LV}\delta} \tag{2}$$

where Young's law has been used to replace the combination of interfacial tensions by $\cos\theta_Y=(\gamma_{SV}-\gamma_{SL})/\gamma_{LV}$. For a liquid droplet of permittivity ε_1 immersed in a second dielectric liquid of permittivity ε_2 we would expect $(\varepsilon_l-1)\rightarrow(\varepsilon_1-\varepsilon_2)$.

Equation (2) is a modified form of Young's law applicable to interface localized L-DEP, which we refer to as dielectrowetting in recognition of the contact angle, θ_e, dependence on the voltage squared. It is similar in form to the electrowetting modified Young's law, but with the ratio of the substrate permittivity to substrate dielectric thickness, ε_r/d, replaced by the ratio of liquid permittivity (minus vapor value) to electric potential penetration depth, $(\varepsilon_l-1)/\delta$. This reflects the change from charging of the solid-vapor interface as a liquid front advances (electrowetting) to polarization of dipoles within a boundary layer of the solid-liquid interface by a non-uniform field (L-DEP) originating from the substrate. Therefore an exponentially decaying electric field penetrating into a dielectric liquid will enhance wetting, reducing the contact angle and so drive a droplet to spread

The equations for electrowetting and dielectrowetting can be brought together in a single notational form by using the value of voltage, V_{TH}, at which complete wetting, i.e. $\cos\theta(V_{Th})=1$, is predicted to occur[23],

$$\cos\theta_e(V_o)=\cos\theta_e(0)-\left[\cos\theta_e(0)-1\right]\left(\frac{V_o}{V_{Th}}\right)^2 \tag{3}$$

Equation 3 provides a more experimentally determined form of the contact angle dependence on the voltage, by using the initial measured contact angle before a voltage is applied, $\theta_e(0)$, and an experimentally deduced threshold voltage, V_{Th}, at which complete wetting, $\theta_e(V_{Th})=0^o$, is expected to occur. This equation is valid for both electrowetting and dielectrowetting and has the advantage of gathering device and liquid parameters into the experimentally determined $\theta_e(0)$ and V_{Th}.

In principle, dielectrowetting can provide an alternative or complementary approach to electrowetting for generating liquid lenses using dielectric liquids. To create liquid lenses which operate in an analogous manner to electrowetting, the challenge is to design suitable structures to create electric fields which decay exponentially with distance from the substrate. One method of doing so in a quasi-two-dimensional geometry is to use co-planar fabricated interdigitated electrodes. In such a case, the electric field decays exponentially with distance above the electrodes, although there is also a modulation of the electric field in the direction across the electrodes. A comparison of traditional electrowetting to dielectrowetting is given in table 1.

Table 1. Comparison of electrowetting to dielectrowetting

Consideration	Electrowetting	Dielectrowetting
Physical mechanism	Capacitive charging of the solid-liquid interface *via* ions in a uniform electric field.	Polarization of dipoles close to the solid-liquid interface *via* a non-uniform electric field.
Liquid	Conducting liquid (typically water + salt).	Dielectric liquid (typically a non-conducting oil).
Voltage source	DC or low frequency AC.	DC or AC (high frequency if the liquid is conducting).
Electrodes	Contact to liquid normally required (see ref. 24 for a contact-less form).	No contact to liquid required.
Electrode structure	Sandwich (involving a solid insulator and sensitive to defects/pin-holes).	Co-planar (surface microfabricated electrodes) or other.
Saturation	Contact angle saturates.	No saturation. Film formation is possible.

2.3 Film and thin film regimes

To produce a non-uniform electric field originating from the substrate, consider a co-planar set of interdigitated electrodes with width and gap between electrodes of, d (Fig. 2 Top view). From Poisson's equation, the solution for the potential in a semi-infinite dielectric liquid is of the form $V(x,z)\sim\cos(kx)\exp(-kz)$, so that the decay length is related to the electrodes by $\delta=4d/\pi$.[19,21,25] Confinement of the liquid to motion along the electrodes occurs due to the variation in voltage across successive electrodes in the x-direction. As a voltage is applied, the liquid will spread along the electrodes and a side profile view taken perpendicular to the electrodes will show a change in contact angle consistent with eq. (2) (Fig. 2a). This is the quasi two-dimensional approach to droplet wetting in the thick film regime with $h<<\delta$.

We now consider $h\rightarrow\delta$ due to a substrate with a set of more widely spaced electrodes, and hence increasing δ, or due to spreading the droplet with increasing applied voltage, V_o, so that the maximum droplet height reduces, or due to depositing a thin liquid film through some other means. In this case, the upper liquid-vapor interface comes within the range of the decaying electric potential. Therefore changes in the shape of the liquid-vapor interface, and hence its surface free energy, become possible, rather than just changes in solid-liquid contact area; this can balance changes in L-DEP energy and a static wrinkle appears on the upper surface of the liquid film (Fig. 2b).

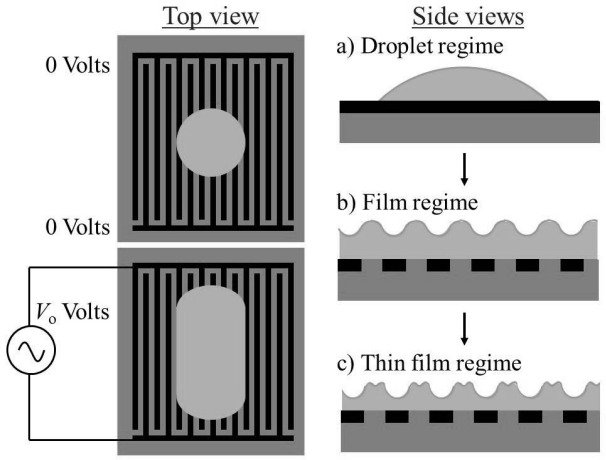

Figure 2. Evolution from droplet to film and thin film regimes as the electric field penetration depth increases.

The energies involved in changes at the liquid-vapor interface can be explained using detailed calculations or a simple scaling argument[21,25]. The equilibrium shape is then found by minimizing the total energy change with respect to changes in wrinkle amplitude, *A*,

$$A \propto \left(\frac{\varepsilon_o (\varepsilon_l - 1) V_o^2}{\gamma_{LV}} \right) \exp\left(\frac{-4h}{\delta} \right) \tag{4}$$

If the liquid is in the very thin film regime, the assumption that the potential from the electrodes along the *x*-direction is a simple cos(*kx*) form breaks down due to charge accumulation along the edges of each electrode finger; the shape of the liquid-vapor interface of the liquid film then follows the near field shape reflecting higher harmonics within the electric field (Fig. 2c). Calculations of the higher frequency components can be performed and the relative amplitudes of components calculated[21].

3. EXPERIMENTAL RESULTS

3.1 Droplets and dielectrowetting

To confirm the validity or otherwise of eq. (2), we conducted droplet experiments using 1, 2 propylene glycol on a substrate fabricated with parallel planar interdigitated electrodes of finger width and gap spacing of 80 μm. In the data shown a hydrophobic 2 μm thick SU-8 film was also used on top of the substrate to prevent accidental damage to the device. The effect of this layer when using a dielectric liquid is to cause a capacitive division of the voltage so that the effective voltage in the liquid is reduced compared to that applied to the interdigitated electrodes[18]. Applying a 10 kHz peak-to-peak voltage causes the liquid to spread along the direction of the electrodes as can be seen from the top and side views in the lower inset of Fig. 3.[18] The side view across the electrodes reveals a droplet cross-section shape from which a contact angle can be measured. This angle decreases with increasing voltage and increases with decreasing voltage with a small degree of hysteresis and a clearly visible cross-over from advancing to receding angle as the voltage goes from increasing to decreasing (Fig. 3). The upper inset in Fig. 3 shows the dependence of $\cos\theta$ on the square of the voltage, V_o^2, and demonstrates a clear linearity. This curve mirrors the relationship observed for electrowetting although in this case the experiment uses a dielectric liquid without any contacting electrodes. The lowest contact angle achieved here was limited by the finite length of the interdigitated electrodes and not saturation effects; in principle smaller angles can be achieved, but the theory only applies whilst $h >> \delta$ remains valid.

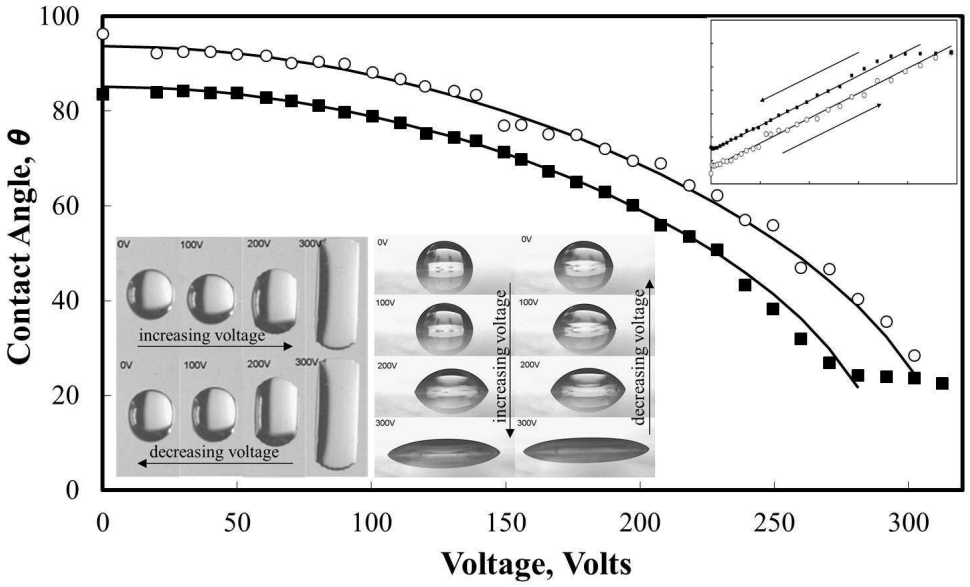

Figure 3. Control of contact angle for stripe shaped droplets of 1,2 propylene glycol on a substrate with interdigitated electrodes. The lower inset shows top and side view images for a range of voltages. The upper inset shows a test of eq. 2. [Data from reference 18].

These results show a quasi two-dimensional, i.e. stripe, form of droplet control, but the theory should be applicable to axisymmetric three-dimensional droplets. Such droplets and lenses using circular interdigitated electrodes have been reported by other researchers[15,16], although we do not believe an interface localized form of L-DEP and a wetting interpretation has been provided other than in reference 18.

3.2 Films and amplitude phase grating

In a separate set of experiments we used a device with an electrode pitch of 20 μm on which a small volume droplet of 1-decanol as deposited[19]. Every other stripe in the electrode array was biased with a 20 kHz a.c. voltage and the interdigitated stripes between them were earthed. When the voltage was increased above a threshold voltage, a uniform film of thickness 3 μm was achieved. Further increases in the amplitude of the voltage caused a static undulation ("wrinkle") to appear at the liquid-vapor interface consistent with eq. (4). The period of the wrinkle matched the electrode pitch with the peaks and troughs parallel to the electrode fingers. Since the wrinkle is static and has an amplitude determined by the applied voltage, a polarization insensitive amplitude-programmable phase diffraction grating can be created in either transmission or reflection mode. The concept for transmission mode is shown in Fig. 4 together with an inset (top left corner) showing the pattern of different order diffractive peaks observed on the screen as light at λ=543 nm is transmitted through the film of oil. Measurements of the intensity of the zero-, first- and second-order diffractive peaks are also shown[19].

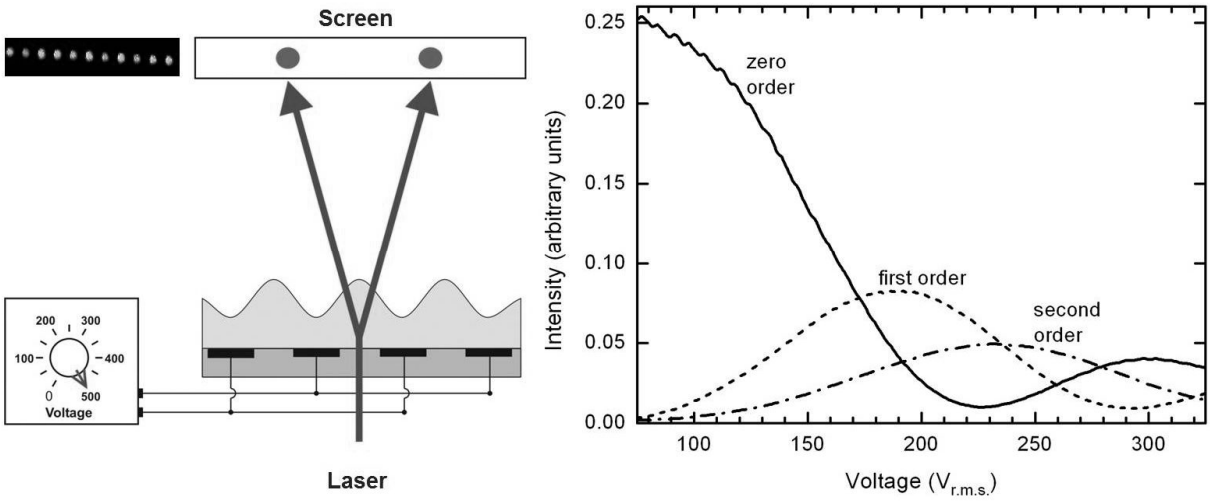

Figure 4. Amplitude phase grating in transmission mode (λ=543 nm) using a 3 micron thick film of 1-decanol. [Data from reference 19].

Further experiments demonstrated that the transient response of the first-order peak for switch-on and switch–off was rapid with a wrinkle of amplitude A=0.36 microns on films of average thickness 14-20 microns requiring 35-49 microsecond (switch-on) and 79- 108 microsecond (switch-off).[19] The speed of the switch-on and switch-off (relaxation) is because the liquid phase-grating only uses a change of the liquid-vapor interfacial shape and does not involve the movement of a contact line.

We also imaged the shape of the liquid-vapor interface using a Mach-Zehnder interferometer for different average thicknesses of a hexadecane oil film (0, 9.0, 16.5 and 30.0 microns) on a device with a 240 micron pitch and a 2 micron capping layer of SU-8. Figure 5 shows the evolution of the profile as the r.m.s voltage is increased from 275 V to 550 V (profiles at different thicknesses have been offset for clarity). A schematic of the expected electric field patterns due to charge accumulation at the edges of electrodes has been included in the figure (after Feldmann & Henaff[26]). This shows how higher order Fourier components matching the near field electric field pattern become evident as the thickness of oil decreases (see also reference 27).

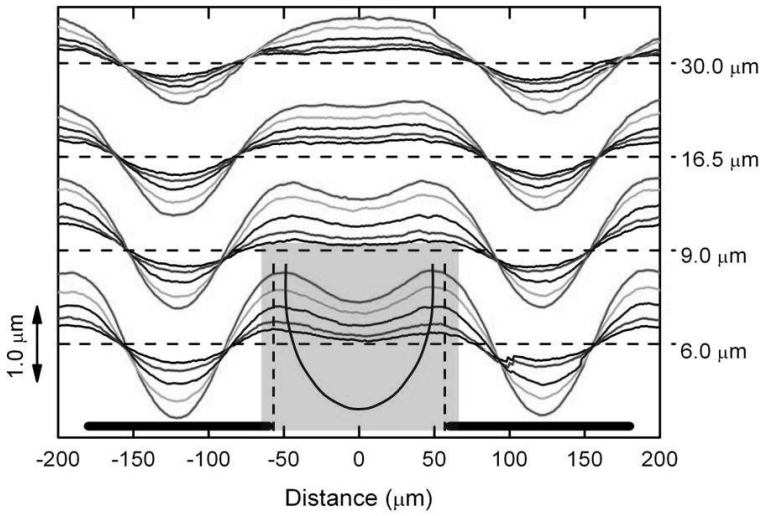

Figure 5. Measured profiles at a hexadecane oil–air interface for films of hexadecane at different average thicknesses induced by the action of a non-uniform electric field profile on a 240 micron pitch interdigitated device. A schematic of the expected electric field profile due to charge accumulation along the edges of the electrode fingers has been included. [Data from reference 19].

An interesting variation on the idea of a voltage programmable liquid amplitude phase grating is to use the ability to monitor and control the diffractive peaks to create an optimized solid amplitude phase grating. Wells et al[28] have shown that using a UV curable resin as the liquid film it is possible to continuously and *in situ* adjust the optical diffraction pattern using the voltage whilst UV curing. This allows the creation of a fully cured solid phase grating with a particular voltage-selected tailored optical property, such as a suppression of the zero transmitted order at a given wavelength.

4. CONCLUSIONS

In this paper we have outlined a theory for L-DEP driven (dielectro-) wetting and shaping of liquid-vapor and liquid-liquid interfaces. The approach uses the concept of localizing the net effects of liquid-dielectrophoresis forces to solid-liquid and liquid-vapor (or liquid-liquid) interfaces. The dielectrowetting equation for droplets is similar in form to the electrowetting equation, but replaces the ratio of the permittivity of a solid insulator to its thickness by the ratio of the liquid permittivity (minus 1) to an electric potential penetration depth. Our experimental realization of these ideas effectively transforms from a sandwich structure device approach to an in-plane one using co-planar interdigitated electrodes. The dielectrowetting approach removes requirements common in electrowetting for a solid insulator, direct electrical contact to the liquid and a conducting liquid. We have also outlined the conditions under which the L-DEP forces shape the liquid-vapor (or liquid-liquid) interface and shown that static undulations ("wrinkles") may be obtained and that these can be used to create voltage programmable amplitude phase gratings, which are fast in switching and polarization independent.

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